# **Liquid Lead-Bismuth Materials Test Loop**

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#### Abstract

We designed and built the Liquid Lead-Bismuth Materials Test Loop (MTL) to study the materials behavior in a flow of molten lead-bismuth eutectic (LBE). In this paper we present a description of the loop with main components and their functions. Stress distribution in the piping due to sustained, occasional and expansion loads is shown. The loop is designed so that a difference of  $100^{\circ}$ C can be attained between the coldest and the hottest parts at a nominal flow rate of 8.84GPM. Liquid LBE flow can be activated by a mechanical sump pump or by natural convection. In order to maintain a self-healing protective film on the surface of the stainless steel pipe, a certain concentration of oxygen has to be maintained in the liquid metal. We developed oxygen sensors and an oxygen control system to be implemented in the loop. The loop is outfitted with a variety of instruments that are controlled from a computer based data acquisition system. Initial experiments include preconditioning the loop, filling it up with LBE, running at uniform temperature and tuning the oxygen control system. We will present some preliminary results and discuss plans for the future tests.

#### I. INTRODUCTION

Liquid lead-bismuth eutectic is considered as prototype target and coolant for the Advanced Accelerator Applications (AAA) Project. It is an alloy 44.5% lead and 55.5% bismuth with the melting temperature of 123.5°C and boiling temperature of 1670°C. This liquid s relatively low melting point and high boiling point in addition to good heat transfer properties make it a very good candidate for a coolant. Relatively large neutron yield and low neutron capture cross section make it an attractive target. Combining the target and coolant roles in one material allows for a simpler target design. Liquid spallation source also eliminates some of the structural damage problems associated with solid targets.

Lead-bismuth has been used successfully as a coolant in submarine nuclear reactors in Russia since 1950 s. There has not been any work done in the US on lead-bismuth since the 1950 s. The reason for that was that in the beginning of the lead-bismuth research structural materials oxidation in liquid lead-bismuth caused significant problems. Russian scientists, however, determined that an oxide film on the surface of stainless

steel protects it from lead-bismuth corrosion. Formation and longevity of this protective film depends on oxygen concentration in the liquid metal. In order to use liquid lead-bismuth in AAA facility we need to know how to control corrosion of structural materials. That is the main goal of the liquid lead-bismuth Materials Test Loop (MTL)

We have designed and MTL to study the thermohydraulic and corrosive behavior of liquid lead-bismuth eutectic. The main goals of this facility are:

- Study of the long-term corrosive effects of liquid lead-bismuth on materials;
- Study of the oxygen control system in the liquid leadbismuth flow;
- Study the natural convection flow in a liquid leadbismuth system;
- Study of liquid lead-bismuth coolant properties.
   MTL is shown on the drawing on Figure 1.
   MTL was designed by Los Alamos National

Laboratory team members in cooperation with Institute of Physics and Power Engineering (IPPE) of Obninsk, Russia.

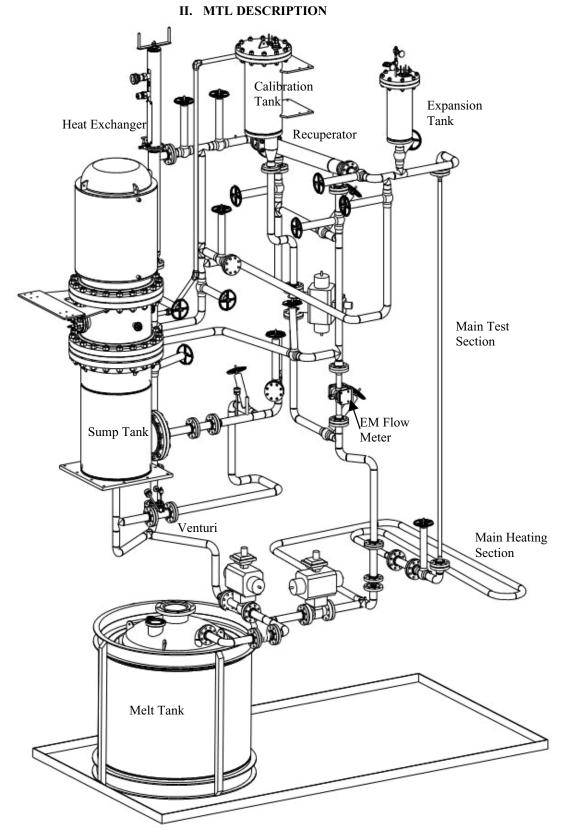


Figure 1. Drawing of Material Test Loop (MTL).

MTL is shown on Figure 1. It is a closed loop consisting of a pump, piping, heat exchangers, and tanks. During operation lead-bismuth is melted in the Melt Tank and transferred by gas pressure into the Sump Tank. A centrifugal pump submerged in the liquid metal in the Sump Tank circulates the fluid through the loop. After leaving the Sump Tank, liquid lead-bismuth travels up to the Recuperator's shell side, where fluid s temperature is increased by 50°C. Magnetic flow meter is placed on the long vertical pipe leading from the Recuperator's shell side to the heated section at the bottom of the loop. Band heaters cover the next five horizontal tubes. There the fluid s temperature is raised another 50<sup>o</sup>C. Then the liquid goes up through a narrower vertical Test Section (1in nominal diameter) and through the tube side of the Recuperator where its temperature is reduced by 50°C. After leaving the Recuperator, the fluid flows to the Heat Exchanger where its temperature is again reduced by 50°C. The fluid leaves the Heat Exchanger through the bottom outlet, goes down through the vertical pipe, turns and returns to the Sump Tank through the bottom inlet. Several pipes are built into the loop to allow bypass of the Recuperator, Heat Exchanger or the Sump Tank. The temperature changes shown in this paragraph are nominal for the design flow speed of 1m/s in the test section or about 6kg/s mass flow rate.

The pump used in MTL is a standard mechanical pump manufactured for lead smelting industry. It is a centrifugal pump with an 8.5in impeller. The impeller is submerged in liquid lead-bismuth during operation. It is driven by a 25 horse power electric motor and is capable of 58GPM maximum flow in the loop. The motor is coupled to the impeller by a long vertical shaft. The pump was built by LaBour Pumps.

The recuperator is a standard shell and tube heat exchanger where both the hot and the cold fluids are liquid lead bismuth at different temperatures. The Heat Exchanger is a special IPPE design. It consists of several

concentric tubes with water as the cooling fluid. Water is separated from the loop fluid by an annulus filled with lead-bismuth. This intermediate fluid can be moved up and down inside the annulus by moving the inside cylinder that works as a piston. When brought all the way up it leaves the intermediate lead-bismuth at the bottom of the outer cylinder thus reducing the heat exchanging capacity of the apparatus to minimum. When the piston is lowered to the very bottom the intermediate fluid is pushed up into the annulus thus increasing the heat exchanging area and the heat exchanger capacity. The piston is moved by turning a screw handle at the top of the apparatus.

All components of the loop are built of standard 316 stainless steel, which is one of the materials to be tested for its interaction with lead-bismuth. MTL also has a test section where coupons of various other materials can be placed for testing in the lead-bismuth flow.

Approximately 8000lb (3400kg) of lead-bismuth is used in the loop. Lead-bismuth eutectic was purchased from Ney Smelting & Refining Co. Inc. Independent chemical analysis confirmed eutectic composition: 55.8% Bismuth and 43.7% lead. All other components are less than 0.032% by weight.

A photograph of the MTL is shown on Figure 2. The loop is surrounded by an enclosure. At the time this photograph was taken the top part of the enclosure was removed. One can see the tape heaters, the thermocouples and the spring hangers supporting the loop.

MTL was designed to maximum temperature of  $550^{0}$ C. Russian experience showed that it is possible to run lead-bismuth systems up to this temperature with oxygen control. Lead-bismuth target designs considered for AAA applications will run at lower temperatures. We intend to run corrosion tests at different constant temperatures for long periods of time such as 3000 hours and 5000 hours.

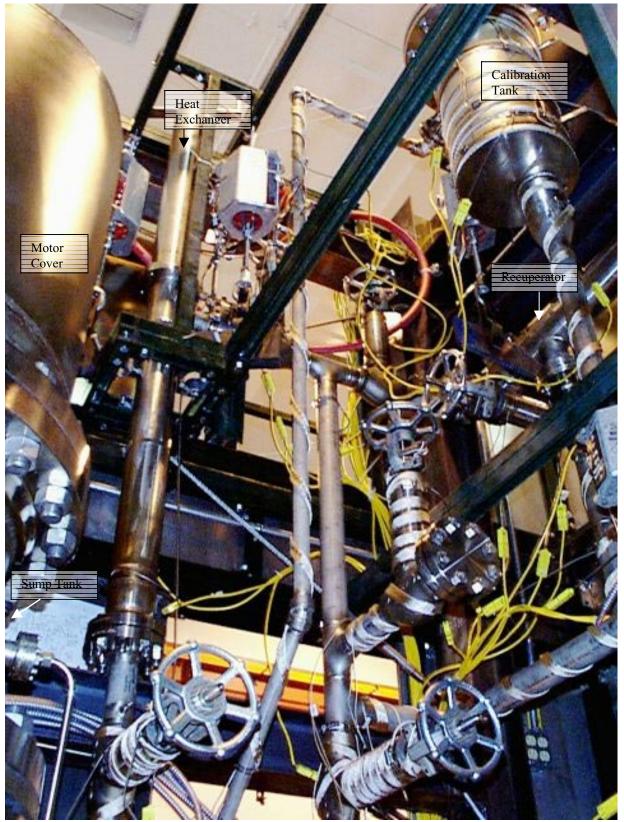


Figure 2. Materials Test Loop. Sump Tank is visible on the left, Heat Exchanger next to ST, Calibration Tank is at the right top corner, and Recuperator is underneath CT, in the shadow.

### III. STRESS ANALYSIS

MTL was designed to minimize the stresses due to gravity, internal pressure, thermal expansion and seismic loads. AutoPIPE 6.0<sup>1</sup>, additional programming and hand calculations were used for the stress analysis. AutoPIPE is a commercial code for piping stress analysis. Figure 3 shows the schematic representation of the loop in AutoPIPE with calculated maximum stress distribution due to thermal expansion indicated in color.

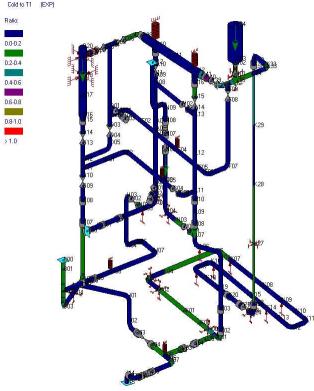


Figure 3. MTL stress distribution due to thermal expansions at maximum temperatures (from AutoPIPE 6.0 analysis).

Figure 3 shows the resulting thermal expansion stresses at maximum nominal temperatures in the loop. That means that the hottest part is the narrow vertical test section that is at  $550^{\circ}$ C, while Sump Tank outlet is at  $450^{\circ}$ C.

Stress calculations due to other loads were performed as well. Gravity loads include the weight of lead-bismuth, the pipe and all of the components.

Seismic loads were calculated according to Uniform Building Code 97<sup>2</sup> rules. Los Alamos National Laboratory requires this Code to be used to determine seismic loads. We determined the seismic load value is 0.2\*W, where W is the weight of the component to which the load is applied. This value is based on the local seismic zone 2B, the type of soil underneath the building and the type of facility. Seismic loads are applied

horizontally. The resulting stress is compared with the allowable stress for occasional loads.

By careful selection of supports, their placement and use of bellows we reduced the stresses to below allowable stresses in stainless steel 316 pipe according to ASME B31.1 Power Piping Code<sup>3</sup>. The stresses in the piping are below allowable for other intermediate temperatures and when the loop is empty.

We used flanges in the MTL for removable short test sections of pipe, to connect drain valves, bellows and other parts. These are standard 300lb flanges made of stainless steel 316. We needed to minimize stresses in the flanges imposed by the loop weight, thermal expansion and internal pressure. After analysis of the nature of stresses in flanges we determined that the initial preload on the flange will be limited to 30ft\*lb of torque on every bolt. We also planned a pressure test to check that the flanges are tight.

# IV. OXYGEN CONTROL SYSTEM

Liquid lead-bismuth reacts with some of the components of stainless steels and, thus, causes corrosion. Scientists at IPPE, Obninsk, Russia, discovered that if an oxide film is allowed to form on the steel surface it prevents corrosion. This protective film consists mostly of steel components oxides and it is based on Fe<sub>3</sub>O<sub>4</sub>. The main purpose of the MTL and its first goal is to implement and to practice operating a system that would maintain this protective film.

The oxygen control system consists of oxygen sensors, gas injection devices and filters.

We have designed and built sensors that measure oxygen content in liquid lead-bismuth<sup>4</sup>. They are based on already existing automotive oxygen sensors. The main part of the automotive oxygen sensor is a doped zirconia ceramic conical probe, see Figure 4. This ceramic is a solid electrolyte that is permeable to oxygen ions at temperatures above 350°C. The minimum temperature was determined in prior bench-top experiments with oxygen sensors of this design<sup>4</sup>. The probe is mounted inside a specially designed cylinder so that it is immersed into the liquid lead-bismuth when the device is mounted on the pipe. Inside the ceramic probe contains a reference, in this case liquid bismuth in equilibrium with its oxide. Voltage is developed due to the difference between the oxygen concentration on either side of the ceramic. Relationship between voltage and oxygen concentration was derived from formulas available in literature and from separate experiments. A drawing of the oxygen sensor design is shown on Figure 4.

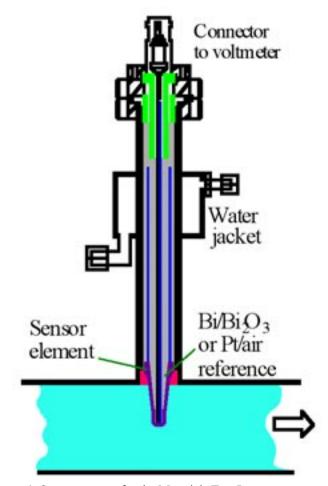


Figure 4. Oxygen sensor for the Materials Test Loop.

The oxygen sensor may allow some liquid leadbismuth travel up its container tube. To prevent leadbismuth from reaching the connectors and leaking outside, a freeze plug will be formed by cooling an area of the tube with water an solidifying the liquid metal.

Since the reference is a liquid metal, the oxygen sensor has to be mounted vertically. Oxygen sensor with air a reference is also being considered. It has to be open to the ambient air, however, which makes it more problematic because that creates a leak path for the lead-bismuth.

There are five oxygen sensors placed around the loop. We measure oxygen content at the pump outlet and inlet, before and after the main heating section and in the Sump Tank's cover gas volume.

The ceramic probe can allow oxygen ions pass through only at temperatures higher than 350°C. That temperature became the minimum operating loop temperature during testing.

In order to maintain the protective oxide film on the stainless steel surface a certain amount of oxygen has to be sustained in lead-bismuth. An oxygen sensor can determine the amount of oxygen we have and a gas injection device can introduce additional oxygen or hydrogen to reduce the amount of oxygen. On the other hand, hydrogen reacts with oxygen and, hence, reduces its amount in the loop. The gas injection device is mounted in a bypass leg of the loop near the Sump Tank. It is a venturi constriction with two small holes for gas inlet. Gas inlet line is connected to two small holes in a venturi constriction. Mass control valves control the flow of gas in and out of the system.

The gas inlet is equipped with level sensors similar to the level sensors in the tanks (see part V). If lead-bismuth reaches the bottom level sensor we can increase the gas inlet flow and push the liquid metal out of the gas inlet tube. A freeze plug prevents lead-bismuth from leaking through the gas inlet pipe.

A differential pressure transducer measures the pressure drop in the venturi. We use this measurement to determine the flow speed through this bypass. The bypass line is connected to the main loop piping via a globe valve. This valve can regulate the flow through the gas injection venturi.

A mesh filter placed at the Sump Tank inlet is meant to catch larger pieces of oxides and other debris carried by the liquid metal.

### V. INSTRUMENTATION

#### V.I. Instruments and devices

The Materials Test Loop is equipped with pressure transducers, thermocouples, an Electro-Magnetic flow meter, level sensors, water flow meters and oxygen sensors. Other equipment includes the pump, heaters and actuated valves.

Standard thermocouples and pressure transducers are use, except the pressure transducers in the lead-bismuth flow have to be mounted away from the pipe to reduce the temperature near the measuring part. Heaters are placed on the tubes connected to the pressure transducers to make sure that they do not get cold enough to solidify lead-bismuth.

Level sensors are steel rods hanging from the top of a tank. When liquid metal reaches a level rod it closes the circuit between the level sensor and the tank wall. Voltage reading at the sensor tells the computer that the level is reached. We designed and built the level sensors specifically for this application. Preliminary tests with water and lead-bismuth confirmed their reliability and convenience.

Electro Magnetic Flow Meter works on the magnetic induction principle. When a conducting fluid, such as liquid metal, flows through a perpendicular magnetic field, it induces an electric current perpendicular both to the flow and to the magnetic field. Thus a permanent magnet is placed around the pipe and two electrodes are welded to the pipe opposite to each other and on a line

perpendicular to the line between the magnet poles. The electrodes are exposed to the liquid metal for more precise measurement. The EM Flow Meter we are using on the loop was designed and built by IPPE, Russia and it is similar to the Flow Meters they use on their liquid metal loops. There are theoretical expressions that relate the measured voltage to the flow speed, but with liquid metal many factors can alter this relationship. We are going to calibrate the EM Flow Meter using a Calibration Tank (see Figure 1).

Oxygen sensors were custom designed as explained in the part IV and in more detail in [4].

We installed a standard centrifugal mechanical pump to propel the liquid lead-bismuth around the loop. Calculations show that the maximum speed that can be developed by the pump in the MTL is 57GPM. The pump is driven by a 25hp motor with a variable drive. Thus, we can run at different flow speeds.

The main heating section is at the bottom of the loop just before the narrow vertical test section. Horizontal pipes are covered with band heaters that all together provide about 60kW of heat. Trace heaters are used everywhere else.

#### PRESS TO START/STOP PROGRAM Pump Enable Current time Output Interval (sec) LBE in Melt Tank 16:55:52 Main Gas System Presets Intermidiate Data T101 26:24 T102 26:30 T103 25:00 T104 84:53 T105 24:97 T109 25.44 18 Pump Speed Control (Hz) T201 T202 26.18 25.40 0.00 T216 25.00 () 0.00 04 25.09 0.03 Pump speed (RPM) 20.0 30.0 ∰ **(0.00** 0.00 0.00 Pump Motor Temperature F203 25.14 250 500 750 T138 T139 T140 25.00 84.21 25.33 T131 26.04 HTR 525 T902 88.07 T132 25.68 T145 24.46 24-Sump Tank PT201 rate Control 0.60 -0.21 \$0.00 0.00 HTR T175 25.13 522A T176 24.94 HTR 514 HTF 512 T146 24.46 18 -0.00 T171 25.15 HTR T172 25.10 \$ 521 T141 24,62 # 1014D 0.00 T147 24.84 11 = T118 85,74 HTÄ 0.00 0.00 T134 25.63 HTR 509A T127 24.89 PT102 T148 25.05 HTR 2.06 HTF 0.00 HTR 518 17153 25.00 17154 25.00 T124 25.00 T116 . 0.00 HTR 0.00 5054 HTR 509H 0.00 **₩ ₩ 0.00** T121 25.04 HTR T161 24.91 0.00 520 HTR 0.00 506 T179 24.65 T180 23.57 T228 T229 24.43 24.49 T114 # E08508 T106 24.98 HTR 503 T112 24.83 0.00 T122 25,28 T119 25.23 0.00 T120 25.52 T163 25.18 T164 24.95 T904 24.58 0.00 527 HTB T181 25.00 T182 25.00 T136 25.08 0.00 FT101 T113 23,61 0.00 T187 0.00 24.79 HTR20-2 T208 24.75 HTR 501A HTR 517A HTR 510A T198 23.93 MT Goal Temp Level T199 24.44 MT Heating Rate Status and Warnings MT is full at 32" 0.00 10.82 -0.04 ST is empty CT is empty ET is empty Required pressure at PT201 is below atmosphericl Yent Value FV203 and He fill value FV201 will close! 0.00 3.00 Required pressure at PT202 is below atmospheriol Vent Valve FV204 and He fill valve FV202 will closel Required pressure at PT202 is below atmospherio!

# V.II. Data Acquisition and Control System

Figure 5. DAC program front panel. T\*\*\* indicates thermocouples, PT\*\*\* - pressure transducers, OS\*\*\* - oxygen sensors, FV\*\*\* - actuated valves, LT\*\*\* - level sensors, HTR\*\*\* - heaters and FT101 is the magnetic flow meter.

A computer based Data Acquisition and Control (DAC) system reads the data collected by the instruments and allows the operator to control the pump, heaters and actuated valves. DAC also includes automatic control algorithms.

DAC system is written in LabVIEW, a data acquisition language developed by National Instruments. National Instruments hardware is used for data input/output. A picture of the DAC front panel is shown on Figure 5.

Examples of the DAC system s automatic functions include overtemperature and overpressure protection. The loop was designed for the maximum temperature of 550°C. The DAC program will shut down the pump and drain lead-bismuth into the Melt Tank if it detects temperatures approaching maximum anywhere in the loop. Some of the points on the cold side of the loop have maximum temperatures of 450°C and the program accounts for that. Detecting high pressure in a tank will first cause a gas vent valve to open in an effort to reduce pressure. If that does not work DAC program will shut down the pump, drain the loop. The program drains the loop by opening actuated valves at the bottom of the loop and allowing the fluid to flow down into the Melt Tank.

There are many complex control situations when the program initiates automatic action.

During the first experimental stage we will learn how to operate the oxygen control system by controlling its mass control gas valves and reading the oxygen sensors. We will determine various values such as heat input from the pump, real Heat Exchanger capacity, maximum flow speed, etc. We will tune the DAC program to maintain pressures and temperatures within acceptable intervals. We will learn how to use the Heat Exchanger.

At the initial stages the MTL will not be operated unattended, but we are planning to run long term experiments in the future. Based on what we learn during the first stage the DAC system will be improved to include more automatic controls so that the MTL can be run unattended.

# VI. CURRENT AND PLANNED TESTING

The MTL has undergone several pressure tests with water under pressures of up to 120psig. Two leaks were detected in the gas fittings. They were fixed. One of the loop flanges was deemed likely to leak and replaced with a welded joint. Total gas leak rate in the MTL is about 3.65 psi/day or 6.2%/day.

Pressure test also allowed us to practice the fluid transfer from the Melt Tank to the Sump tank and to the loop. As a result we know the best gas and fluid valves positions during the transfer. We also know approximate pressures during transfer.

We tested the gas control system injecting helium into the venturi inlet with water in the loop. We were

able to control the pressures in the Sump Tank and the Expansion Tank. We also successfully maintained the water level inside the gas injector preventing it from going up the gas inlet tubes.

Next operation was filling up the Melt Tank with lead-bismuth. A small tank was attached to the top of the Melt Tank via a pipe with a valve and a filter. Leadbismuth ingots were loaded into the small tank by hand and melted. When completely melted the valve separating the Melt Tank will be opened and the liquid metal will drain into the Melt Tank. This procedure will continue until the top level in the Melt Tank is reached. Loading lead-bismuth in this manner will prevent excessive amount of lead and bismuth oxides getting into the Melt Tank. The oxides melt at higher temperature than lead-bismuth eutectic so they were removed from the liquid before the transfer into the Melt Tank. This operation was necessary because some of the leadbismuth containers allowed water to leak in and the ingots surfaces became oxidized.

First lead-bismuth transfer into the MTL and circulation with the pump will be done at about 350°C. We will first run the loop without using the main heaters and the Heat Exchanger. During this time we will test the gas injection and regulation system. We will calibrate the Magnetic Flow Meter.

Next, heating rates, Heat Exchanger and Recuperator temperature decrease at different flow speeds will be determined. We will also determine the thermocouples that best represent the temperature at the heating zones.

When we are confident in the MTL operations and the DAC system is updated and tested for unattended operations, we will put steel samples into the test section and start the first long term run.

In the future we would like to utilize the flexibility of design that we built into the MTL. We want to run the loop with a liquid metal flow driven by natural convection. We also want to test liquid lead-bismuth coolant properties on prototype geometries for AAA targets or any other designs.

# VII. CONCLUSION

The Materials Test Loop is a valuable tool in studying properties of liquid lead-bismuth. We will be able to learn the corrosive effects of lead-bismuth on standard US stainless steels. We will also learn how to control the corrosion and improve steel surface. Some of this liquid metal s cooling properties can be determined even during the initial testing, but even more valuable data will be derived from tests with specific geometries.

The MTL was built to accommodate lead-bismuth eutectic, but the same design and testing principles can be utilized for other liquid metals. Since liquid metals are primary coolants considered for AAA applications the MTL presents a useful facility for future work.

# VIII. REFERENCES

- 1. AutoPIPE 6.1, Rebis, 2000.
- 2. Uniform Building Code 1997.
- 3. ASME B31.1, Power Piping, 1998
- T. DARLING, N. LI, Oxygen Concentration Measurement in Liquid Pb-Bi Eutectic, AccAPP-ADTTA 01 meeting.